Indirect spin interaction via the phonon field. Exact results

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For a spin system interacting with a phonon system, a procedure is introduced for a transition to a new representation via successive canonical transformations the sum of whose generators must satisfy the Frohlich condition. It is found that the generators can be so chosen that the transition to the new representation can be carried out mathematically exactly and explicitly. Averaging of the transformed Hamiltonian over the equilibrium phonon states yields a compact expression for the indirect-spin-interaction operator in terms of the phonon field in all approximations in the spin-phonon interaction.

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§1. INTRODUCTION

Let us consider subsystems of spins and phonons described by the Hamiltonian $H_0 = H_{0s} + H_{0ph}$:

$$H_{0s}=\hbar\omega_0\sum_{j=1}^{N} S_j^z, \qquad H_{0ph}=\hbar\sum_{q}\omega_q(b_q^+b_q+{}^1/_2), \tag{1}$$

and let these subsystems be connected by the interaction

$$H_{sph}=i\hbar\sum_{j=1}^{N}\sum_{q} G_\alpha{}^q S_j^\alpha e^{i\mathbf{q}\mathbf{r}_j}(b_q-b_{-q}{}^+), \tag{2}$$

where ω_0 and ω_q are the Zeeman frequency and the frequency of an acoustical phonon with wave vector **q** and polarization σ: $q = (\mathbf{q},\sigma)$; b_q^+ and b_q are the phonon second-quantization operators, S^α is the α component of the spin vector $\mathbf{S}_j$; $G_\alpha^q = G_\alpha(|\mathbf{q}|/M\hbar v)^{1/2}$ is the spin-phonon coupling constant, $\mathbf{r}_j$ is the radius vector of the spin $\mathbf{S}_j$; M is the mass of the crystal; v is the velocity of sound waves in the crystal.

In the description of purely magnetic properties of a spin system, it is desirable to have a Hamiltonian that does not depend on the vibrational degrees of freedom. This Hamiltonian can be obtained with the help of a molecular-field-type approximation, which in our case will consist of averaging over the vibrational degrees of freedom with an equilibrium density matrix.

However, in the case of direct averaging of the total Hamiltonian of the system $H = H_0 + H_{sph}$, which leaves only the operator of the Zeeman spin energy H_{0s}, the spin-system properties due to coupling with the phonons are completely lost.

A way out of this difficulty is to change to a new representation in which the molecular-field approximation provides additional spin-Hamiltonian terms that depend on the constant G_α. These terms will be proportional to the powers of the constant G_α, beginning with the second, since averaging with the equilibrium density matrix causes the terms that are linear in G_α to vanish. To prevent loss of the principal contribution from the operator H_{sph}, which is proportional to G_α, we must carry out the transition to $\tilde{H}$ in such a way that the terms that are linear in G_α contained in it are mathematically exact. Thus, we arrive at the procedure suggested by Fröhlich[1] in the problem of the interaction of electrons via a phonon field.

According to Ref. 1, the transition to the new representation is effected with the help of the canonical transformation $\tilde{H} = e^{-iL}He^{iL}$, where the Hermitian generator of the representation L is determined from the condition

$$H_{sph}+i[H_0, L]=0, \tag{3}$$

because of which terms that are linear in the coupling constant G_α are absent from $\tilde{H}$.

In the realization of this procedure, great difficulties arise with the calculation of the operator $\tilde{H}$, because of which one usually confines oneself to terms of order G^2_α only, which bear information on the indirect interaction of the spins via the phonon field.[2,3] In the next section, we shall change to a new representation, which is different from the Fröhlich representation and which, satisfying the condition (3), nevertheless leads to an exact expression for the operator $\tilde{H}$.

The calculation of all the approximations in the spin-phonon interaction is not only desirable but also necessary. The reason is that the potentials of indirect interaction in all orders, beginning with the second, contain divergent denominators of the type $\omega^2_0 - \omega^2_q$ and perturbation theory is not applicable to resonant phonons located near ω_0.

§2. CHOICE OF THE NEW REPRESENTATION

To simplify the following exposition, we limit ourselves here to the special case of the interaction (2) with $G^q_z = 0$.[1)] Then by rotation of the coordinate system about the z axis (the constant magnetic field) we can, without further loss in generality, reduce the operator (2) to

$$H_{sph}=i\hbar\sum_{i=1}^{N}\sum_{q} G_x{}^q S_j{}^x e^{i\mathbf{q}\mathbf{r}_j}(b_q-b_{-q}{}^+). \tag{4}$$

We now subject the total Hamiltonian of the system H to two successive canonical transformations with generators L_1 and L_2:

$$\tilde{H}=e^{-iL_2}e^{-iL_1}He^{iL_1}e^{iL_2}=H_0+H_{sph}+\frac{i}{1!}[H_0, L_1]+\frac{i}{1!}[H_{sph}, L_1]$$
$$\ldots+\frac{i}{1!}([\mathrm{H}_0, L_2]+[H_{sph}, L_2]+\frac{i}{1!}[[H_0, L_1], L_2]+\ldots)+\ldots \tag{5}$$

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and require that terms that are linear in the coupling constant G^q_x be absent from the operator $\widetilde{H}$. This imposes on the generators L_1 and L_2 the condition: $H_{sph} + i[H_0, L] = 0$, $L = L_1 + L_2$, which is identical with (3). Solving Eq. (3) in the interaction representation, we can find an explicit expression for the operator L:

$$L=\sum_{j=1}^{N}(S_j^x\xi_j+S_j^y\eta_j),$$

$$\xi_j=-\sum_q G_x^q\omega_q z_q e^{i\mathbf{q}\mathbf{r}_j}(b_q+b_{-q}^+), \tag{6}$$

$$\eta_j=-i\omega_0\sum_q G_x^q z_q e^{i\mathbf{q}\mathbf{r}_j}(b_q-b_{-q}^+),$$

where $z_q^{-1} = \omega^2_0 - \omega^2_q$. Equation (6) leaves the leeway in the choice of the operators L_1 and L_2, and this turns out to be enough for us to effect the transition to the operator $\widetilde{H}$ in explicit form and exactly. This procedure will be accomplished below by choosing

$$L_1=\sum_{j=1}^{N}S_j^x\xi_j,\qquad L_2=\sum_{j=1}^{N}S_j^y\eta_j.$$

It is easy to show that a canonical transformation with such generators as L_1 and L_2 of the total Hamiltonian of the system H can be carried out mathematically exactly. For example, if we transform the operators b_q and b_q^+, the series (5) will contain only two terms:

$$e^{-iL_1}b_qe^{iL_1}=(e^{-iL_1}b_q^+e^{iL_1})^+=b_q+i\sum_{j=1}^{N}S_j^x[b_q,\xi_j],$$

$$e^{-iL_2}b_qe^{iL_2}=(e^{-iL_1}b_q^+e^{iL_2})^+=b_q+i\sum_{j=1}^{N}S_j^y[b_q,\eta_j].$$

The series (5), in the case of the transformation of the spin operators, can be convoluted into the compact, exact expressions

$$e^{-iL_1}S_j^ze^{iL_1}=S_j^z\cos\xi_j-S_j^y\sin\xi_j,$$

$$e^{-iL_2}S_j^ze^{iL_2}=S_j^z\cos\eta_j+S_j^x\sin\eta_j,$$

$$e^{-iL_1}S_j^ye^{iL_1}=S_j^y\cos\xi_j+S_j^z\sin\xi_j,\qquad e^{-iL_2}S_j^ye^{iL_2}=S_j^y,$$

$$e^{-iL_1}S_j^xe^{iL_1}=S_j^x,\qquad e^{-iL_2}S_j^xe^{iL_2}=S_j^x\cos\eta_j-S_j^z\sin\eta_j.$$

Substituting these expressions in (5), we obtain the exact form of the Hamiltonian H in the new representation[2)] $\widetilde{H} = \widetilde{H}_{0s} + \widetilde{H}_{0ph} + \widetilde{H}_{sph}$:

$$\begin{aligned}
\widetilde{H}_{0s}=\frac{\hbar\omega_0}{2}\sum_{j=1}^{N}\Big\{&(S_j^z\cos\eta_j+S_j^x\sin\eta_j)\\
&\times\Big[e^{i\xi_j}\prod_{k=1}^{N}(\cos a_{jk}+2iS_k^y\sin a_{jk})\\
&+e^{-i\xi_j}\prod_{k=1}^{N}(\cos a_{jk}-2iS_k^y\sin a_{jk})\Big]\\
&+iS_j^y\Big[e^{i\xi_j}\prod_{k=1}^{N}(\cos a_{jk}+2iS_k^y\sin a_{jk})\\
&+e^{-i\xi_j}\prod_{k=1}^{N}(\cos a_{jk}-2iS_k^y\sin a_{jk})\Big]\Big\},\\
\widetilde{H}_{0ph}=H_{0ph}+\hbar\omega_0\sum_{j=1}^{N}&S_j^y\xi_j+\hbar\omega_0^2\sum_{j,k=1}^{N}\sum_q|G_x^q|^2\omega_qz_q^2e^{i\mathbf{q}\mathbf{r}_{jk}}S_j^yS_k^y\\
+i\hbar\sum_{j=1}^{N}\sum_q&G_x^q\omega_q^2z_qe^{i\mathbf{q}\mathbf{r}_j}(S_j^x\cos\eta_j-S_j^z\sin\eta_j)(b_q-b_{-q}^+)\\
+\hbar\sum_{j,k=1}^{N}\sum_q&|G_x^q|^2\omega_q^3z_q^2e^{i\mathbf{q}\mathbf{r}_{jk}}(S_j^x\cos\eta_j-S_j^z\sin\eta_j)\\
&\times(S_k^x\cos\eta_k-S_k^z\sin\eta_k),\\
\widetilde{H}_{sph}=i\hbar\sum_{j=1}^{N}\sum_q&G_x^qe^{i\mathbf{q}\mathbf{r}_j}(S_j^x\cos\eta_j-S_j^z\sin\eta_j)(b_q-b_{-q}^+)\\
+2\hbar\sum_{j,k=1}^{N}\sum_q&|G_x^q|^2\omega_qz_qe^{i\mathbf{q}\mathbf{r}_{jk}}(S_j^x\cos\eta_j-S_j^z\sin\eta_j)\\
&\times(S_k^x\cos\eta_k-S_k^z\sin\eta_k),\\
\mathbf{r}_{jk}=\mathbf{r}_j-\mathbf{r}_k,\qquad &a_{jk}=\omega_0\sum_q|G_x^q|^2\omega_qz_q^2e^{i\mathbf{q}\mathbf{r}_{jk}}.
\end{aligned}\tag{7}$$

The results apply to the case of spin $S = \frac{1}{2}$.

We note that the sum $L = L_1 + L_2$ is the generator of the well-known Fröhlich transformation. The form (6) of the operator L is the same in both cases and is determined by the condition (3) that the terms in H that are linear in G_x vanish. However, the fact that the transition to the new representation proposed here is effected by successive canonical transformations with the generators L_1 and L_2, simplifies this procedure decisively, making possible its exact satisfaction, whereas the transformation with the generator $L = L_1 + L_2$ (Ref. 1) leaves perturbation theory as the only possibility.

The basis of the representation in which the initial Hamiltonians (2) and (4) act is the direct product of the spin and phonon subspaces of the eigenfunctions:

$$\Psi_{\{m\}\{n\}}=\prod_{j=1}^{N}\Psi_{j\{n\}}^{m}=\prod_{j=1}^{N}\chi_j^m|\{n\}\rangle,$$

$$|\{n\}\rangle=\prod_{q=0}^{q_D}|n_q\rangle,\quad \{n\}=n_{q_1}\ldots n_{q_D},\qquad \{m\}=m_1,m_2,\ldots,m_N,$$

where $m_j = \pm\frac{1}{2}$ and n_q are the magnetic quantum number of the spin and the number of phonons of type q, respectively; q_D is the wave vector of the Debye phonon. In the new representation, the basis is made up of the functions

$$\tilde{\Psi}_{\{m\}\{n\}}=e^{-iL_2}e^{-iL_1}\Psi_{\{m\}\{n\}}$$

$$=\prod_{j=1}^{N}[A_j^{\pm}\chi_j^{\pm}\pm B_j^{\pm}\chi_j^{\mp}]\,|\{n\}\rangle=\prod_{j=1}^{N}\tilde{\Psi}^{\pm}_{j\{n\}}, \tag{9}$$

where the phonon operators $A_j^{\pm}$ and $B_j^{\pm}$ are equal to

$$A_j^{\pm}=\cos\frac{\eta_j}{2}\cos\frac{\xi_j}{2}\pm i\sin\frac{\eta_j}{2}\sin\frac{\xi_j}{2},$$

$$B_j^{\pm}=\sin\frac{\eta_j}{2}\cos\frac{\xi_j}{2}\mp i\cos\frac{\eta_j}{2}\sin\frac{\xi_j}{2}$$

and $\chi_j^{\pm}$ are the eigenfunctions of the operator S^2_j corresponding to $m_j=\pm\frac{1}{2}$.

The Hamiltonian (7) as constructed does not contain terms of order G_x. This means that under its influence the probability W of a transition between the states of the new basis $\tilde{\Psi}_{(m)(n)}$ can contain a dependence on G_x only starting with G_x^4. Yet in the initial representation the transition probabilities between the states of the spin system under the action of the spin-phonon interaction H_{sph} (4) are proportional to G_x^2. This is explained by the fact that the basis of the new representation consists of functions which are a mixture of the initial spin states. The probability of admixture of one of the initial states in another new state is a function of G_x, the expansion of which begins with G_x^2.

Actually, the probability of observing the state $\Psi^{\mp}_{j\{n\}}$ in the state $\tilde{\Psi}^{\pm}_{j\{n\}}$ of the new basis is

$$|\langle\Psi^{\mp}_{j\{n\}}|\tilde{\Psi}_{j\{n'\}}\rangle|^2=|\langle\{n\}|B_j^{\pm}|\{n'\}\rangle|^2.$$

Since lim $\tilde{\Psi}^{\pm}_{j(n)}\Psi_{j(n)}$ as $G_x\to0$, the normalized probability of mixing of only spin states on going to the new representation is

$$W_{\pm\mp}{}^j=\sum_{\{n\},\{n'\}}|\langle\{n\}|B_j^{\pm}|\{n'\}\rangle|^2\rho(\{n\}), \tag{10}$$

where $\rho(\{n\})$ is the equilibrium density of the phonon states. In second order in the spin-phonon coupling constant G_x, the probability of mixing of the functions χ_j^- and χ_j^+ is equal to

$$W^j_{\pm\mp(2)}=\frac{3|G_x|^2\omega_0^3}{8\pi\hbar\,dv^5\Gamma}\left(\operatorname{cth}\frac{\hbar\omega_0}{2kT}\pm1\right)$$

$$=A^{(1)}_{\pm\mp}/\Gamma\equiv\frac{\pi\omega_0\theta_{\pm\mp}(T)}{4\Gamma}C,$$

where $C=3|G_x|^2\omega^2{}_0/2\pi^2\hbar dv^5$; d is the density of the crystal; T is the temperature of the phonon reservoir; $A^{(1)}_{\pm\mp}$ are the probabilities of single-phonon emission (absorption) processes for the spin system, and Γ is the total width of the magnetic-resonance line. The parameter Γ enters into the expression for $W^j_{\pm\mp(2)}$ thanks to the use of the following approximation:

$$z_q^2=\frac{1}{(\omega_0+\omega_q)^2(\omega_0-\omega_q)^2}\approx\frac{\pi}{2\omega_0^2\Gamma}\delta(\omega_0-\omega_q), \tag{11}$$

which is necessary for integration of the contribution from the resonant phonons, which we describe by the isotropic Debye model.[3] Taking it into account that the decisive contribution to the magnetic resonance line width is made by the spin-phonon interaction, for which $\Gamma=(\pi\omega_0C/2)\coth(\hbar\omega_0/2kT)$, we obtain

$$W^j_{\pm\mp(2)}=\frac{1}{2}\left(1\pm\operatorname{th}\frac{\hbar\omega_0}{2kT}\right),\qquad W^j_{\pm\pm(2)}=\frac{1}{2}\left(1\mp\operatorname{th}\frac{\hbar\omega_0}{2kT}\right), \tag{12}$$

where $W^j_{\pm\pm(2)}$ is the probability that the spin j remain in the same state when account is taken of the interaction with the phonons.

We can also obtain an exact expression for the probability (10) without approximations in terms of the constant G_x:

$$W_{\pm\mp}{}^j=\frac{1}{2}\left[1-e^{-1}\pm e^{-1/2}\sin\left(\operatorname{th}\frac{\hbar\omega_0}{2kT}\right)\right]. \tag{13}$$

The probabilities (12) and (13) turn out to be independent of the spin-phonon coupling constant. The explanation of this fact is that by condition (11) we exclude from consideration all phonons except the resonant phonons. As is well known, the spin is transformed from the states $\chi_j^{\pm}$ to the states $2^{-1/2}(\chi_j^{\pm}\pm\chi_j^{\mp})$, under the influence of interaction with the resonant classical field. This corresponds to the field-independent transition probability $W^j_{\pm\mp}=\frac{1}{2}$. The only interaction of the spin with the quantized field introduces the sole change that as $T\to0$ only the probabilities of spontaneous transitions remain different from zero, so that the upper spin levels of the energy are frozen out:

$$(W^j_{++(2)}=W^j_{-+(2)}=0).$$

§3. EFFECTIVE SPIN HAMILTONIAN

In correspondence with what was said in Sec. 1, we average the energy operator (7) with the equilibrium phonon-density matrix. After this, the effective Hamiltonian of the spin system can be represented in the form

$$H_{ss}=H_{ss(1)}+H_{ss(2)}+\ldots,$$

where $H_{ss(n)}$ is an operator describing the effective interaction of n spins. In this way, we obtain for the single-spin Hamiltonian

$$H_{ss(1)}=\hbar\omega_0\sum_{j=1}^{N}S_j^z[D_je^{-A(T)}+2A_1(T)e^{-A_1(T)}],$$

$$A(T)=\frac{1}{2}\sum_q|G_x^q|^2(\omega_0^2+\omega_q^2)z_q^2\operatorname{cth}\frac{\hbar\omega_q}{2kT}, \tag{14}$$

$$D_j=\prod_{k=1}^{N}\cos a_{jk},$$

$$A_1(T)=\frac{1}{2}\sum_q|G_x^q|^2\omega_0^2z_q^2\operatorname{cth}\frac{\hbar\omega_q}{2kT}.$$

Thus, the quantity $\hbar\omega_0\delta_j$, where

$$\delta_j=D_je^{-A(T)}+2A_1(T)e^{-A_1(T)}-1, \tag{15}$$

is the shift in the frequency of magnetic resonance due to spin-phonon coupling. If we disregard surface spins, the quantities D_j, and with them the shifts δ_j, do not depend on j. Equation (14) takes into account the contribution to the resonance frequency shift from the spin-phonon interaction (4) in all orders.

We pause to look in detail at the operator $H_{ss(2)}$, which is traditionally regarded as the Hamiltonian of indirect interaction of the spins through the phonon field:

$$H_{ss(2)}=\hbar\omega_0\sum_{j\neq k=1}^{N}\sum_{\alpha,\beta=x,y,z}P_{jk}^{\alpha\beta}S_j^\alpha S_k^\beta, \tag{16}$$

where the nonzero interaction potentials are equal to

$$P_{jk}^{xx}=\frac{1}{2\omega_0}\sum_q|G_x^q|^2\omega_q(2\omega_0^2-\omega_q^2)z_q^2e^{i\mathbf{q}\mathbf{r}_{jk}}[e^{-B_{jk}^+}+e^{-B_{jk}^-}],$$

$$P_{jk}^{yy}=\sum_q|G_x^q|^2\omega_0\omega_q z_q^2e^{i\mathbf{q}\mathbf{r}_{jk}}$$

$$-2e^{-B}\left(D_j\operatorname{tg}a_{jk}-\sum_{l=1}^{N}D_l\operatorname{tg}a_{ll}\operatorname{tg}a_{lj}\operatorname{tg}a_{lk}\right),$$

$$P_{jk}^{zz}=-\frac{1}{2\omega_0}\sum_q|G_x^q|^2\omega_q(2\omega_0^2-\omega_q^2)z_q^2e^{i\mathbf{q}\mathbf{r}_{jk}}[e^{-B_{jk}^+}-e^{-B_{jk}^-}].$$

The parameters of this equation have the following meaning:

$$B_{jk}^{\pm}=\sum_q|G_x^q|^2\omega_q^2z_q^2(1\pm\cos\mathbf{q}\mathbf{r}_{jk})\operatorname{cth}\frac{\hbar\omega_q}{2kT},\qquad B=\frac{B_{jj}^+}{4}.$$

As already noted, the equation for the potentials of indirect two particle interaction (16) were obtained without approximation in the constant G_x. They constitute rather complicated integrals with respect to the phonon wave vectors, which are calculated only approximately. Such a calculation is given in the Appendix, and using its results we obtain the following equations for the contribution of the resonant phonons to the potentials

$$P_{jk}^{xx}=e^{-1}a_{jk}\operatorname{ch}\left(\frac{\sin\rho_{jk}}{\rho_{jk}}\right),\qquad P_{jk}^{zz}=e^{-1}a_{jk}\operatorname{sh}\left(\frac{\sin\rho_{jk}}{\rho_{jk}}\right),$$

$$P_{jk}^{yy}=a_{jk}-2e^{-1/2}D\left(\operatorname{tg}a_{jk}-\operatorname{tg}a_{jj}\sum_{l=1}^{N}\operatorname{tg}a_{lj}\operatorname{tg}a_{lk}\right), \tag{17}$$

$$a_{jk}=\operatorname{th}\frac{\hbar\omega_0}{2kT}\frac{\sin\rho_{jk}}{\rho_{jk}},\qquad a_{jj}=\operatorname{th}\frac{\hbar\omega_0}{2kT},\qquad D=D_j.$$

It appears strange at first glance that the potentials (17) turn out to be independent of the spin-phonon interaction constant and therefore do not vanish in the limit $G_x\to 0$. This is explained by the fact that the transition to the limit in the potentials (17) is not valid, since it was assumed in obtaining them that the spin-phonon contribution to the level width exceeded all the others and thus was bounded below. Just as (A.2), the more general formulas should contain the ratio $C\omega_0/\Gamma$, where Γ is the total magnetic-resonance level width, which as $G_x\to 0$ goes over into the width determined by the other interactions.

In contrast to all the existing calculations of spin-spin indirect interactions through the phonon field, which were always obtained in the approximation of second order perturbation theory in the coupling constant G_x, the result (16) is obtained from the mathematically exact expression for the transformation of the Hamiltonian. We note that since the transformation (5) converts the system to a representation that is different from the Fröhlich representation, the second-order perturbation theory, which is obtained from the result (16), need not coincide with the traditional expression for the indirect interaction, obtained previously in Refs. 2 and 3.

The procedure of successive canonical transformations that we have proposed turned out to be effective. It allows us to solve the same problem in a significantly broader formulation than the result communicated here. Thus, we have obtained an exact equation for the operator $H_{ss(2)}$ in the case of spin-phonon coupling of the form (2). The restrictions to spins $S=\frac{1}{2}$, and also to the two-particle spin-spin interaction in H_{ss} have only a quantitative character. The operators of other multispin interactions can be obtained from the transformed Hamiltonian $\widetilde{H}$ (7), whereas for another spin the Hamiltonian $\widetilde{H}$ itself must be derived anew. Moreover, the results extend automatically to an arbitrary quantum system having a discrete spectrum and interacting with a boson field of arbitrary nature. For example, it can be extended to atoms having optical energy levels and interacting with the electromagnetic field.

However, in the present paper we shall not develop the results that further in these indicated directions, but shall confine our attention to one important problem which arose in the course of our obtaining the exact expression for $H_{ss(2)}$.

§4. DISCUSSION OF THE RESULTS

To obtain the exact expression for the operator $H_{ss(2)}$, a transition was proposed in Sec. 2 to a new representation, with the help of two successive canonical transformations, which were subject to a single condition (3) that determined the sum $L=L_1+L_2$ of the generators of these transformations, and also to a tacit condition that follows from the purpose of our research, namely that the transformation (5) be followed up to conclusion with mathematic rigor. Within the framework of these conditions, the question of the uniqueness of our splitting of L into L_1 and L_2 remains open: one can choose some other pair of operators $L_1'=L_1-\Delta L$ and $L_2'=L_2-\Delta L$ with arbitrary Hermitian ΔL. In particular, setting $\Delta L=L_2$, we obtain the Fröhlich transformation and it is unclear *a priori* which of these representations should be preferred. Moreover, choosing certain L_1 and L_2, we nevertheless have leeway in the sequence of realization of these transformations:

$$\widetilde{H}=e^{-iL_2}e^{-iL_1}He^{iL_1}e^{iL_2}\ \text{и}\ \widetilde{H}'=e^{-iL_1}e^{-iL_2}He^{iL_2}e^{iL_1}. \tag{18}$$

Since $[L_1,L_2]\neq 0$, the transition in Eq. (18) to different representations and the Hamiltonians $\widetilde{H}$ and $\widetilde{H}'$ are generally speaking not equal.

At first glance, it may appear that the indicated arbitrariness does not have serious significance, since the measured physical quantities do not depend on the representation in which their operators are specified. However, in Sec. 3 the Hamiltonian (7) was projected on a spinor subspace by replacing all the phonon operators by their equilibrium mean values at a given temperature. Since such a projection is not an exact procedure, the resultant Hamiltonian operator and hence, the properties of the spin system are generally dependent on the choice of the new representation.

The problem of the correct choice of the operators L_1 and L_2 turns out to be very difficult, since it is impossible to formulate mathematically the restrictions imposed on their form by the condition of solvability of the transformation. There remains the path of formulation of the rules for this choice on the basis of general physical and intuitive considerations.

One of such criteria can be the condition of the invariance of the direction of the spin quantization axis in the new representation. One cannot expect the spin-phonon interaction (2), which describes the fluctuations of the local magnetic field, to be capable of creating local magnetic field with zero mean value, whose direction does not coincide with the quantization axis.

For example, it was possible for us to realize an exact transition to the new representation for two other subdivisions of the operator L that correspond to

$$\Delta L=\sum_{j=1}^{N} S_j^y\xi_j \text{ and } \Delta L=\sum_{j=1}^{N} S_j^x\eta_j.$$

In both cases, after averaging Hamiltonian $\tilde{H}$ over the equilibrium states of the phonons, the spin quantization axis acquired components in the xy plane and was turned through an angle proportional to arctan $(G^2_x/\hbar^2\omega^2_0)$. We have rejected these transformations on this basis and considered transitions to new representations only with

$$L_1=\sum_{j=1}^{N} S_j^x\xi_j, \qquad L_2=\sum_{j=1}^{N} S_j^y\eta_j,$$

which do not change the direction of the quantization axis in the effective spin Hamiltonian H_{ss} regardless of the order of carrying out the transformations with the operators L_1 and L_2.

To choose the order of carrying out the transformations with the generators L_1 and L_2, we made use of the principle of minimum free energy in the new representation. As is well known, the free energy of a system of N spins can be represented in the form

$$F=-kT\ln \operatorname{Sp} e^{-H_{ss}/kT}\approx -kTN\ln\left[\operatorname{ch}\frac{\hbar\omega_0(1+\delta)}{2kT}\right].$$

The last equation is obtained if we take H_{ss} to be only the spin term $H_{ss(1)}$. The shift of the Zeeman frequency in the Hamiltonian $\tilde{H}'$ is equal to

$$\delta'=\delta+e^{-A(T)}\sum_q |G_x^q|^2 z_q \operatorname{cth}\frac{\hbar\omega_q}{2kT}.$$

Taking it into account that the second term of this expression is less than zero, we see that $\delta'<\delta$ and consequently, the free energy has a minimum in the representation with the Hamiltonian $\tilde{H}$, as discussed in detail in this paper.[4]

There is a possibility, within the restricted framework of the second approximation, to illustrate the differences that arise in the potentials of indirect interaction because of the differences between the Fröhlich representation and successive canonical transformations with the generators L_1 and L_2 chosen above. It is seen from the expansion (5) that the second approximation in the spin-phonon interaction takes in the case of the Fröhlich representation the form

$$\tilde{H}_{Fr}^{(2)}=H_0+i[H_{sph},L]-\frac{1}{2}[[H_0,L],L]$$
$$=\sum_{j\neq k=1}^{N}\sum_q |G_x^q|^2\omega_q z_q e^{i\mathbf{q}\mathbf{r}_{jk}}S_j^xS_k^x, \tag{19}$$

since in the other case considered

$$\tilde{H}^{(2)}=H_0+i[H_{sph},L]-[[H_0,L_1],L_2]$$
$$-{}^1/_2[[H_0,L_1],L_1]-{}^1/_2[[H_0,L_2],L_2]$$
$$=\tilde{H}_{Fr}^{(2)}+\hbar\omega_0\sum_{j\neq k=1}^{N} a_{jk}(S_j^xS_k^x-S_j^yS_k^y). \tag{20}$$

We average the operators (19) and (20) over the equilibrium states of the phonons and transform in the isotropic model of Debye from summation over the phonon wave vectors to integration. The potential of the operator (19), which contains a pole of first order z_q, is calculated in the sense of its principal value:[3]

$$(P_{jk}^{xx})_{Fr}$$
$$=C\left(\frac{\omega_D}{\omega_0}\right)^3\left[-\frac{\sin R_{jk}}{R_{jk}^3}+\frac{\cos R_{jk}}{R_{jk}^2}-\frac{\pi}{2}\left(\frac{\omega_0}{\omega_D}\right)^2\frac{\cos\rho_{jk}}{R_{jk}}\right]. \tag{21}$$

The calculation of the potential a_{jk} in the right-hand side of Eq. (21) is given in the Appendix:

$$P_{jk}^{xx}=(P_{jk}^{xx})_{Fr}-P_{jk}^{yy}, \qquad P_{jk}^{yy}=-\frac{\pi}{2}C\frac{\omega_0}{\Gamma}\frac{\sin\rho_{jk}}{\rho_{jk}}. \tag{22}$$

In addition to the contribution in (22) from the resonant phonons, the potential P_{jk}^{yy} contains also the contribution (A.1) of the nonresonant phonons, which, as is shown in the Appendix, is significantly smaller.

The second approximation obtained with the same pair of canonical transformations but in reversed sequence [see Eq. (18)] yields exactly the same potential (22).

In conclusion, we note that the procedure suggested by us for the calculation of the spin-phonon interaction in all approximations does not remove the divergences either in the second or in the higher terms of the series (5). As a consequence, the criterion of applicability of perturbation theory is not satisfied in the resonance region. For example, calculating for the potential P_{jk}^{xx} the ratio of terms of fourth order in the interaction constant to terms of second order we obtain

$$P_{jk}^{xx(4)}/P_{jk}^{xx(2)}=-(B_{jk}^{+}+B_{jk}^{-})\approx -2.$$

This confirms the necessary of summing all order of perturbations theory, which was in fact done in this work.

APPENDIX

We calculate the integrals encountered in the potentials of the indirect-interaction Hamiltonian (16). For example, we calculate the following integral for the phonons in the approximation of the isotropic model of Debye:

$$a_{jk}=\omega_0\sum_q|G_x{}^q|^2\,\omega_q z_q{}^2 e^{i\mathbf{q}\mathbf{r}_{jk}}$$

$$=\frac{3V|G_x|^2\omega_0}{(2\pi)^3M\hbar v}\int_0^{q_0}dq\,q^2\int_{-\pi}^{0}d\theta\sin\theta\int_0^{2\pi}d\varphi\,q\omega_q\frac{e^{(iqr_{jk}\cos\theta)}}{(\omega_0{}^2-\omega_q{}^2)^2}$$

$$=\frac{C}{\rho_{jk}}\int_0^{\omega_D}d\omega_q\,\omega_q{}^3\frac{\sin(\omega_q r_{jk}/v)}{(\omega_0{}^2-\omega_q{}^2)^2}\equiv(a_{jk})_{nonR}+(a_{jk})_R,$$

where $C=3|G_x|^2\omega_0/2\pi^2\hbar\rho v^5$ is a dimensionless constant, V is the volume of the crystal, and $r_{jk}=|r_{jk}|$. We divide the entire integration region into two parts: resonant $[\omega_0-\Gamma/2,\omega_0+\Gamma/2]$ where Γ is the total width of the magnetic resonance line, and the entire remaining nonresonant region. Since the maximum contribution to the integral is made in the nonresonant region by the high-frequency phonons, we can set approximately $z_q\approx\omega_q^{-2}$ in the integral and carry out the integration over the entire phonon spectrum, since now the narrow resonance band has no singularities,

$$(a_{jk})_{nonR}=C\left(\frac{\omega_D}{\omega_0}\right)\frac{\mathrm{Si}(R_{jk})}{R_{jk}},\quad R_{jk}=\frac{\omega_D}{v}r_{jk}=\frac{r_{jk}}{a},\tag{A.1}$$

where a is the lattice constant. In the resonance region, the approximation (11) is valid, and gives

$$(a_{jk})_R=\frac{\pi}{2}C\frac{\omega_0}{\Gamma}\frac{\sin\rho_{jk}}{\rho_{jk}},\quad\rho_{jk}=\frac{\omega_0}{v}r_{jk}.\tag{A.2}$$

Both parts of the function $a_{jk}(R_{jk})$ in fall off in like fashion with distance and the ratio of their constants is

$$(a_{jk})_R/(a_{jk})_{nonR}\approx\omega_0/\Gamma\gg1.$$

Thus, the contribution from the resonant phonons to the integral is decisive. In this connection, we present below the potentials (16) calculated in the resonance region:

$$P_{jk}{}^{xx}=\theta^{-1}(T)\frac{\sin\rho_{jk}}{\rho_{jk}}[e^{-B_{jk}{}^+}+e^{-B_{jk}{}^-}],$$

$$P_{jk}{}^{zz}=-\theta^{-1}(T)\frac{\sin\rho_{jk}}{\rho_{jk}}[e^{-B_{jk}{}^+}-e^{-B_{jk}{}^-}],\tag{A.3}$$

$$P_{jk}{}^{vv}=2\left[\theta^{-1}(T)\frac{\sin\rho_{jk}}{\rho_{jk}}-e^{-B}\left(D_j\,\mathrm{tg}\,a_{jk}-\sum_{l=1}^{N}D_l\,\mathrm{tg}\,a_{ll}\,\mathrm{tg}\,a_{lj}\,\mathrm{tg}\,a_{lk}\right)\right],$$

$$a_{jk}=2\theta^{-1}(T)\frac{\sin\rho_{jk}}{\rho_{jk}},\quad a_{jj}=2\theta^{-1}(T),\quad\theta(T)=4\Gamma/\pi\omega_0C,$$

$$B_{jk}{}^{\pm}=2\theta^{-1}(T)\,\mathrm{cth}\frac{\hbar\omega_0}{2kT}\left[1\pm\frac{\sin\rho_{jk}}{\rho_{jk}}\right],\quad B=\theta^{-1}(T)\,\mathrm{cth}\frac{\hbar\omega_0}{2kT}.$$

For the parameters $A(T)$ and $A_1(T)$ in the expression for the shift of the magnetic-resonance frequency we have

$$A(T)=2\theta^{-1}(T)\,\mathrm{cth}\frac{\hbar\omega_0}{2kT},\quad A_1(T)=\theta^{-1}(T)\,\mathrm{cth}\frac{\hbar\omega_0}{2kT}.\tag{A.4}$$

The expressions (A.3) and (A.5) are simplified appreciably if, as in Sec. 2 we assume in the calculations of the transition probabilities $W_{\pm\mp}{}^j$, that the magnetic-resonance line width is determined by the spin-phonon interaction. Then

$$\Gamma=A_{+-}^{(1)}+A_{-+}^{(1)}=\frac{\pi\omega_0C}{2}\,\mathrm{cth}\frac{\hbar\omega_0}{2kT},$$

where $A_{\pm\mp}^{(1)}$ are the probabilities of one-phonon processes of emission-absorption by the spin system.[4] In this case, $\Theta(T)=2\coth(\hbar\omega_0/2kT)$ and the parameters $A_1, A, B_j^{\pm}$ and B lose the temperature dependence, which is preserved only in a_{jk} and D_j.

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